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Regional Metamorphism of Ultramafic Rocks in the Central Alps: Parageneses in the System CaO-MgO-SiO₂-H₂O

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With 6 figures and 1 table in the text

Abstract

A sequence of seven critical mineral assemblages has been recognized in regional metamorphosed ultramafic rocks of the Lepontine and Rhetic Alps. This sequence is consistent with experimentally determined and calculated equilibria within the system CaO-MgO-SiO₂-H₂O.

Index assemblages in the ultramafic rocks are compared with index minerals or assemblages in rocks of pelitic, basic, and carbonate composition; conditions of metamorphism range from the prehnite-pumpellyite facies to the upper amphibolite facies. Tremolite-olivine schists are restricted to medium grades of metamorphism; at low grades diopside coexists with serpentine and at high grades the join diopside-enstatite is stable.

I. Introduction

Mineral assemblages corresponding to nearly all the divariant regions in P-H₂O-T space determined for the system MgO-SiO₂-H₂O by BOWEN and TUTTLE (1949) and GREENWOOD (1963) have been recognized in the regionally metamorphosed ultramafic rocks of the Central Alps. The component CaO creates additional recognizable parageneses whose limits are the univariant equilibria corresponding to the lower and upper stability limits of forsterite plus tremolite. The introduction of CO₂, liberated by prograde reactions in neighbouring

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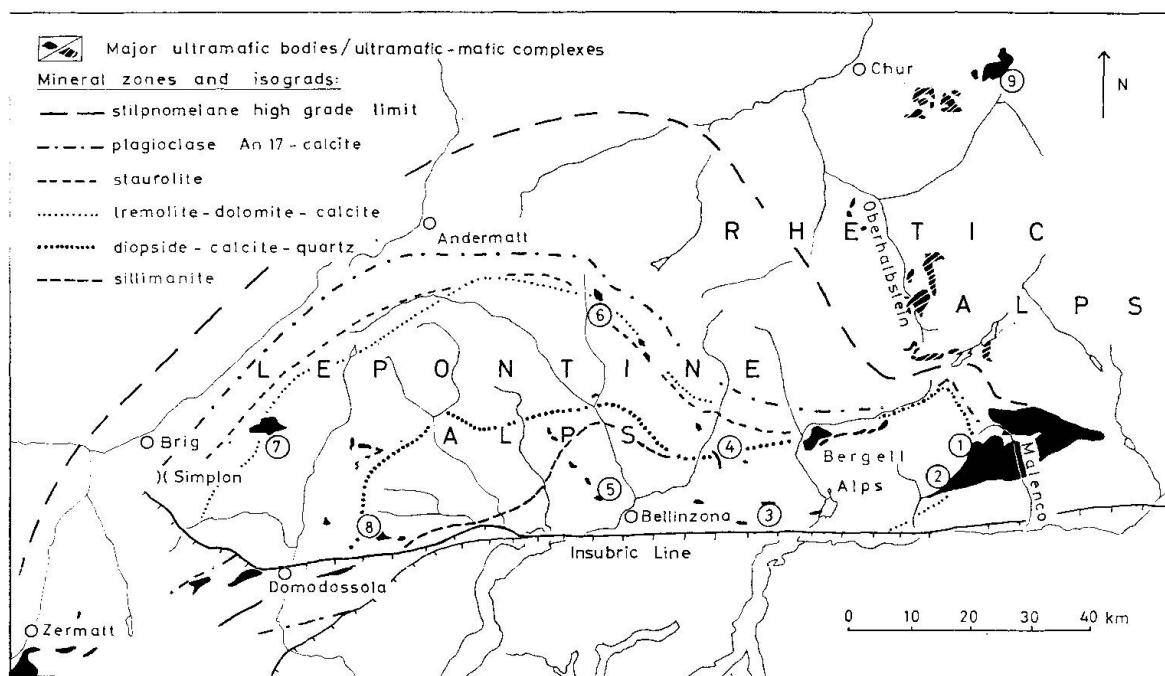


Fig. 1. Distribution of metamorphosed ultramafic rocks in the Central Alps. Circled numbers designate localities mentioned in text: 1 Alpe Zoca, Val Ventina; 2 Corni Bruciati, Val Preda Rossa; 3 Borgo, Val Dargeno; 4 Alpe di Mea and Val Cama; 5 Alpe Arami; 6 Cima Giu, Val Carassina; 7 Geisspfad; 8 Swiss border, Centovalli; 9 Totalp near Davos.

calcareous schists (Bündnerschiefer), has locally produced magnesium silicate plus magnesite rocks. These will be the subject of a later paper.

Bodies of ultramafic rock occur widely in the Central Alps (Fig. 1). They range in size from more than 100 km² to masses so small that their original chemical identity has been lost through metasomatic contact reaction. In the vast majority of cases they have undergone regional metamorphism of alpine age, with the result that structures and metamorphic grade are shared in common with the surrounding schists and gneisses. These latter include calcareous and pelitic rocks of the Bündnerschiefer, metadolomites and marbles, all of which are Mesozoic in age, together with a variety of siliceous gneisses. At lower metamorphic grades, it is clear that the ultramafic rocks belong to a Mesozoic ophiolite association (STAUB, 1922; CORNELIUS, 1935; PETERS, 1963; BEARTH, 1967; DIETRICH, 1969).

As a preliminary to the present discussion, some generalizations based on an extensive sampling and literature review are assembled below.

1. The ultramafic rocks have undergone a very thorough metamorphism of alpine age, in which crystallization outlasted penetrative deformation.
2. Mineral and textural homogeneity (i. e. absence of zoning, mineral relics, and grain-to-grain variability, and the presence of homogeneous mosaic and/or lepidoblastic fabrics), together with repeated occurrences of simple mineral

assemblages, are all factors strongly suggestive that equilibrium conditions were attained during recrystallization of the ultramafic rocks.

3. Silica-poor assemblages (i. e. forsterite-bearing) are widely preserved. Excluding obvious monomineralic metasomatic contact zones and such volatile species as H_2O , CO_2 , and O_2 , the metamorphism was largely isochemical.

4. At metamorphic grades above the stability limit of antigorite, mineral assemblages representing crystallization under different H_2O activities tend to occur together in many outcrops. The association may be a zoned one, or one involving a veining relationship (TROMMSDORFF and EVANS, 1969b).

5. A relatively simple mineral chemistry permits close comparison with equilibrium relationships in the synthetic system $MgO-SiO_2-H_2O$. The influence of the component FeO is minimal because, either before or during metamorphism, there was a tendency for oxidation to take place, with the result that iron/magnesium ratios in the silicate minerals cover a range which extends to much lower values than typical for unaltered ultramafic rocks (TROMMSDORFF and EVANS, 1969a). The component CaO is often present and gives rise to either tremolite or diopside (see below). In the P-T and compositional range encountered, the components Al_2O_3 and ferric iron are responsible for ubiquitous chlorite and magnetite respectively.

II. Experimental and calculated equilibria in the system $CaO-MgO-SiO_2-H_2O$

Listed in Table 1, in order of increasing temperature, are reactions of significance in the metamorphism of ultramafic rocks. Aside from the last reaction (no. 10) the list does not extend beyond the range of equilibrium conditions encountered in the Central Alps.

Table 1. *Reactions in the "ultramafic" system: $CaO-MgO-SiO_2-H_2O$*

Serpentine	+	2 Quartz	=	Talc + H_2O	(1)
Serpentine	+	Brucite	=	2 Forsterite + 3 H_2O	(2)
5 Serpentine	+	2 Diopside	=	Tremolite + 6 Forsterite + 9 H_2O	(3)
5 Serpentine			=	6 Forsterite + Talc + 9 H_2O	(4)
9 Talc	+	4 Forsterite	=	5 Anthophyllite + 4 H_2O	(5)
Anthophyllite	+	Forsterite	=	9 Enstatite + H_2O	(6)
7 Talc			=	3 Anthophyllite + 4 Quartz + 4 H_2O	(7)
Anthophyllite			=	7 Enstatite + Quartz + H_2O	(8)
Tremolite	+	Forsterite	=	5 Enstatite + 2 Diopside + H_2O	(9)
Tremolite			=	3 Enstatite + 2 Diopside + Quartz + H_2O	(10)

(1), (2): JOHANNES, 1968. (5), (6), (7), (8): GREENWOOD, 1963. (10): BOYD, 1959.
(3), (4), (9): calculated, this work.

The locations of these equilibria in PH_2O-T space, as determined experimentally by the authors cited at the foot of Table 1, or calculated in this work (see below), are shown in Fig. 2. Reactions (3) and (9) provide a lower and

upper temperature limit to the occurrence of tremolite in forsterite-bearing rocks.

Reaction (3). This equilibrium is closely related to (2) and (4) (Table 1). From equilibrium (2) $\times 3$, it is obtained by the addition of the vapour-absent equilibrium:



Alternatively, from (4) it can be obtained by adding another vapour-absent equilibrium:



Except at very high pressures, ΔG (11) is positive, and ΔG (12) is negative. This is indicated by both thermochemical data (KING et al., 1967, ROBIE and WALDBAUM, 1968) and petrographic observation, namely, the occurrence of serpentine + diopside (PETERS, 1963, 1968) and the incompatibility in meta-dolomites of diopside + talc (TROMMSDORFF, 1966; SKIPPER, 1967). Hence curve (3) must be located between (2) and (4) (Fig. 2). At two kilobars, thermochemical data (from SHARP, 1962, and the sources cited above) would suggest equilibrium (3) occurs about 24° C above (2) and about 26° C below (4). Curves (2) and (4) are separated by 53° C at 2 kilobars when calculated directly from the same thermochemical data. The experimental curves for (2) and (4) appear to be more widely spaced than this (JOHANNES, 1968; YODER, 1966; KITAHARA et al., 1966), although equilibrium (4) has still not been satisfactorily reversed at pressures of a few kilobars. Until (3) has been located experimentally, it will be assumed provisionally to be midway between (2) and (4). Fig. 2 shows JOHANNES' (1968) version of (2), which is only 20° C lower than (2) by calculation (see also KING et al., 1967, Fig. 4), a calculated version of (4), and curve (3) placed midway between these two.

Enthalpy and low temperature entropy data for chrysotile rather than antigorite were used in the calculation of equilibria (2), (3) and (4), whereas antigorite is the polymorph that commonly occurs at high temperature in nature (DEER et al., 1962, p. 186). This error may well be as large as those arising from uncertainties in the tabulated thermochemical data (from the oxides): ± 40 ° C for (2) and (4), ± 80 ° C for (3).

Reactions (11) and (12) are possible univariant equilibria that occur on the phase diagram at very high pressures. They are generated when (3) intersects (2) or (4) respectively (only one intersection can be stable, of course). Possible topological relations cannot be unambiguously depicted, however, because (11) and (12), together with (13), another topologically possible vapour-absent equilibrium (which would give an upper pressure limit to serpentine):



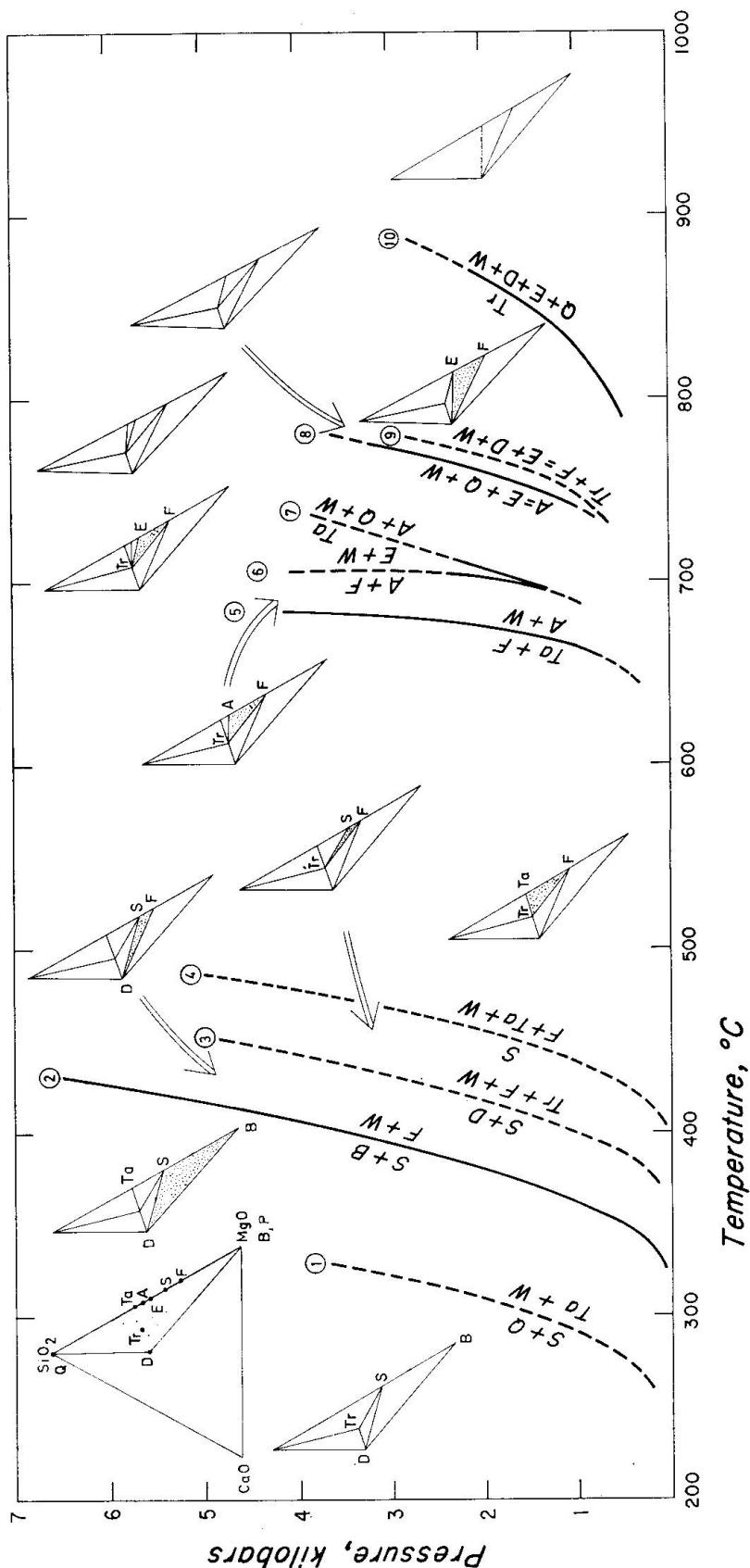


Fig. 2. Pressure-temperature plot of phase relations in the subsystem $MgO\text{-}CaMgSi_2O_6$ (diopside composition)- $SiO_2\text{-}Fe^{2+}H_2O = P_{total}$. Chemographic diagram is a segment of the triangle $SiO_2\text{-}CaO\text{-}MgO$ as shown above left. Diagnostic assemblages recognized in the Alps are indicated by dotted pattern. Solid curves represent experimentally determined equilibria, broken curves represent calculated equilibria. A = anthophyllite, B = brucite, D = diopside, E = enstatite, F = forsterite, P = periclase, Q = quartz, S = serpentine, Ta = talc, Tr = tremolite, W = H_2O .

intersect at another invariant point, located very roughly at 35 kilobars and 450°–500° C, which may then be above or below (2), (3), and (4). Uncertainties resulting from the use of current high temperature thermochemical data and 298° K, 1 atm. volume data at these pressures are very considerable.

Experimental data to 30 kilobars on equilibria (2) and (4) (KITAHARA et al., 1966) give no indication of the existence of (13) at high pressures, despite a ΔV of -6.9 cm^3 at 298° K and 1 atm. for (13). Thus, the vapour-absent invariant point, containing (11), (12), and (13), if it exists at all, may be at much higher pressures than 35 kilobars.

Reaction (9). This equilibrium can be derived from (10), the breakdown reaction of tremolite (BOYD, 1959), by adding the equilibrium:



Thermochemical data (ROBLE and WALDBAUM, op. cit., KING et al., op. cit.) and common petrographic experience indicate a negative ΔG for (14). Although the uncertainties in the tabulated data are not large, reaction (9) involves only one mole of water; thus the calculated curve is uncertain by as much as $\pm 50^\circ \text{ C}$, plus the uncertainty in curve (10). The mean data suggest that equilibrium (9) occurs 70° C (at 500 bars) to 95° C (at 2000 bars) below (10). Thus (9) falls rather close (Fig. 2) to the upper limit of stability of anthophyllite (GREENWOOD, 1963). A direct experimental determination of (9) would be highly desirable.

III. Field occurrence of critical assemblages

Data from over eighty localities of ultramafic rocks from the Central and Rhetic Alps permit recognition of a sequence of mineral associations which are critical with respect to progressive metamorphism. The criteria used to establish the alpine age of the metamorphic ultramafic assemblages are those used and described in an earlier paper (TROMMSDORFF and EVANS, 1969a). It is found that reactions observed to take place in the plane $\text{MgO}-\text{SiO}_2-\text{H}_2\text{O}$ as outlined by TROMMSDORFF (1970) can be augmented by reactions involving the calcic phases tremolite and diopside. Fig. 3 gives a diagrammatic summary of the index assemblages within the subsystem diopside-talc-brucite in the context of index minerals or assemblages from other common rocks in the Alps (WENK, 1962, 1970; E. NIGGLI and C. NIGGLI, 1965; TROMMSDORFF, 1966; PETERS 1963; DIETRICH, 1969).

Particularly noteworthy in the low-grade ultramafics (serpentinites) is the widespread occurrence of diopside. It is well developed in the profile Davos-Oberhalbstein-Malenco (Rhetic Alps, upper penninic to lower east alpine nappes), where the conditions of alpine regional metamorphism range from zeolite facies in the north to high greenschist facies in the south. PETERS (1963)

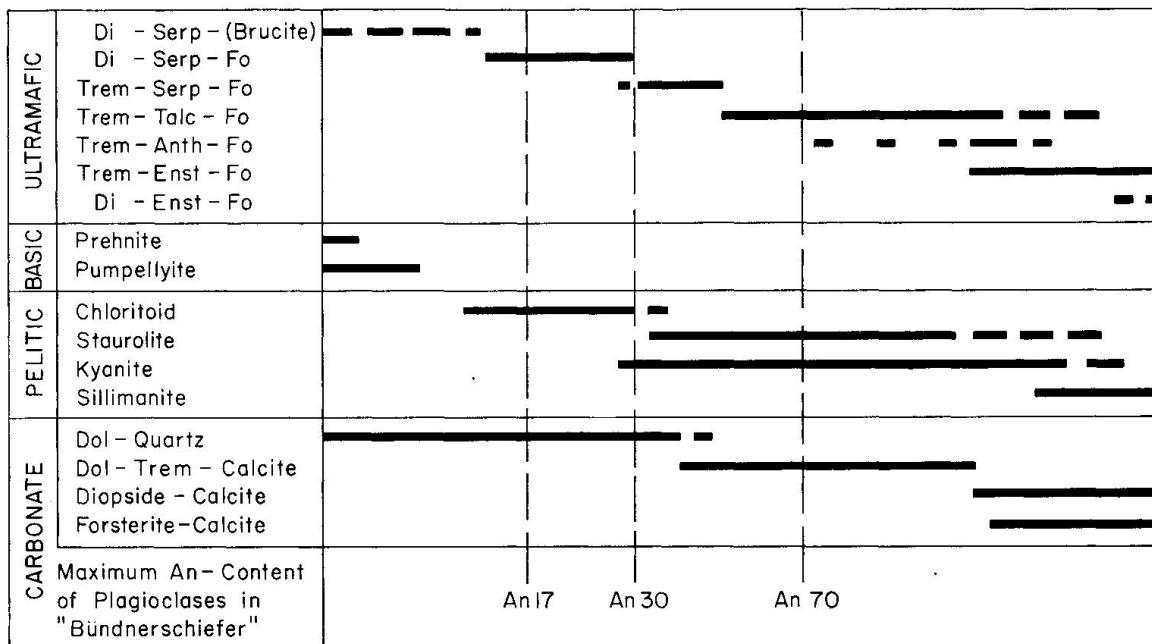


Fig. 3. Index parageneses in ultramafics of the Central Alps in relation to index minerals or parageneses in associated rocks. Low grade data are based mainly on the Oberhalbstein-Malenco profile, data above An 30 on the Lepontine-Bergell area. Broken streaks mark local, full streaks regional occurrences. Data for basic, carbonate and pelitic compositions were compiled after WENK (1962), PETERS (1963), E. NIGGLI and C. NIGGLI (1965), TROMMSDORFF (1966), DIETRICH (1969) and E. NIGGLI (1970).

described postkinematic veins of very pure diopside from the Totalp chrysotile-lizardite-serpentinite at Davos, where the pair prehnite + pumpellyite is stable in metabasaltic rocks. In the adjacent Oberhalbstein area diopside + serpentine (chrysotile-lizardite and antigorite) are common in veins (DIETRICH, 1969) and rocks (MÜLLER, 1959), and in the olivine-antigorite schists of Val Malenco clear alpine diopside granules surround cloudy clinopyroxene reliques (DE QUERVAIN, 1963, PETERS, 1968).

In the following the field evidence for reactions listed in Table 1 and depicted in Fig. 2 will be discussed step-by-step. Because some involve bulk compositions outside the triangle Fo-Di-En, not all the reactions enumerated above are found in the Alpine ultramafics. Indeed at temperatures above reaction (2), rocks lacking olivine are rare and can generally be shown in the field to have suffered some sort of reaction metasomatism.

The change from brucite + serpentine to olivine (+ serpentine) takes place in the Davos-Oberhalbstein-Malenco profile. Brucite occurs in veins in the Oberhalbstein serpentinites, which are olivine-free. At the eastern end of the Malenco serpentinite mass, brucite occurs occasionally in forsterite-diopside antigorite schists (DIETRICH and DE QUERVAIN 1968). Olivine occurs in both veins and rocks in the Malenco serpentinite mass. Along with diopside, it forms

Fig. 4



Fig. 5



Fig. 6

Fig. 4. Diopside - forsterite - antigorite schist, Chiesa, Val Malenco, Italy. PPL, $\times 90$.

Fig. 5. Tremolite - forsterite - antigorite schist, Alpe Zoca, Val Ventina, Italy. PPL, $\times 90$.

Fig. 6. Tremolite - enstatite - forsterite schist, Alpe di Mea, Valle Leggia, Switzerland. PPL, $\times 90$.

a beautiful mosaic texture which seems to be typical for the Malenco antigorite schists (Fig. 4). Thus, there is no doubt that equilibrium (2) occurs within the stability field of the serpentine + diopside assemblage; the latter is apparently stable over most of the Davos-Malenco profile. Formation of new diopside and olivine in antigorite rocks of greenschist environment has also been reported from the Zermatt area (BEARTH, 1967). At least above equilibrium (2), it is clear that antigorite is the stable serpentine polymorph (CORNELIUS, 1935; PETERS, 1968; DIETRICH, 1969; KEUSEN, 1970).

Progressive metamorphism of the olivine-diopside-antigorite schist of Val Malenco can be observed in the contact aureole of the Bergell granite. Tremolite + olivine + antigorite takes the place of diopside + antigorite (reaction (3)) at Alpe Zoca, Val Ventina and at Corni Bruciati, Val Preda Rossa (Fig. 1) within areas no more than 1 km from the granite contact (Fig. 5). Closer to the contact, antigorite reacts, according to (4), to give talc plus more olivine (+ tremolite), and, in the immediate vicinity of the granite contact at Corni Bruciati, this assemblage is locally replaced by anthophyllite + olivine (+ tremolite), following equilibrium (5).

In the Lepontine area the breakdown of the assemblage serpentine + diopside takes place near the staurolite isograd. The replacing assemblage tremolite + olivine has been found in some thin sections of the serpentinite body of Cima Giu, in Val Carassina, Adula Mts., which is located high in the chloritoid zone (EGLI, 1966). Within the staurolite zone the assemblage antigorite-olivine-tremolite is common in the Geisspfad serpentinite in the Simplon area (PREISWERK, 1901) and was first recognized as a metamorphic association by KEUSEN (1970).

Once formed, the pair olivine + tremolite remains stable towards higher metamorphic grades over nearly the whole range of metamorphic conditions in the Lepontine-Bergell area. Olivine + tremolite survives all reactions taking place along the binary join talc-olivine which lead to the formation of anthophyllite and enstatite. Although anthophyllite is quite a common mineral, the assemblage anthophyllite + olivine is not encountered very often, which is consistent with the small temperature difference between equilibria (5) and (6). On the other hand, talc + olivine and enstatite + olivine, both frequently with tremolite, are widely developed in the higher metamorphic zones (Fig. 3). In the sillimanite zone particularly, schists carrying enstatite, olivine, tremolite, and chlorite are very common (DAL VESCO, 1953; KNUP, 1958; BLATTNER, 1965). In the present study this assemblage has been observed at more than 25 localities between the Bergell and the Swiss border in the Centovalli; it commonly forms rocks with a beautiful mosaic texture (Fig. 6).

The assemblage enstatite-diopside-olivine, corresponding to temperatures above (9), is observed in only a few localities, where it survived or grew only under conditions of apparently low H_2O -activity, e. g. at Alpe Arami (GRUBEN-

MANN, 1908, MÖCKEL, 1968) and at Borgo, in Val Darenco, Italy (BLATTNER, 1965). This assemblage is figured in DAL VESCO (1953, Fig. 9).

IV. Conclusions

The sequence of reactions and mineral assemblages identified in metamorphosed ultramafic rocks in the Central Alps is in good agreement with experimental and theoretical data for the system $\text{CaO}-\text{MgO}-\text{SiO}_2-\text{H}_2\text{O}$.

Ultramafic rocks with an appreciable amount of CaO will contain a diopsidic pyroxene at low and high metamorphic grades, and tremolite at intermediate grades. The survival of diallage and growth of new clinopyroxene in serpentinites can be ascribed to the stability of diopside + serpentine + H_2O at low temperatures and pressures. Alternative assemblages, such as tremolite + brucite, occur only at very high pressures (if at all), and tremolite + olivine only at higher temperatures (above (3)). Olivine + clinopyroxene rocks ("wehrellites") have the same field of stability as "dunites" at low temperatures in the presence of H_2O . "Harzburgites", on the other hand, are limited to temperatures above (6) in the presence of H_2O , and "lherzolites" to above (9).

Despite the availability of alumina, in the form of chlorite, the amphibole is invariably tremolite rather than hornblende. The occurrence of the pair tremolite + forsterite in the ultramafics does correspond fairly well to the amphibolite facies, as indicated by assemblages in associated rocks (Fig. 3). Chlorite is quite insensitive to metamorphic grade, occurring from the prehnite-pumpellyite facies to the high amphibolite facies.

Since antigorite is stable to relatively high metamorphic grades – in the Geisspfad mass it is in the staurolite zone (KEUSEN, 1970) – reaction (4) in the case of antigorite may take place at appreciably higher temperatures than in the case of chrysotile.

Isograds in ultramafic rocks can have the same significance as conventional isograds, e. g. those in pelitic rocks, if it can be shown that H_2O activities were uniform, or, better, were equal or close to the maximum possible. Where veins possess the same mineral assemblage as country rock, e. g. Malenco serpentinite, such high H_2O activities may have been fairly general. On the other hand, low H_2O activities seem to be indicated in certain enstatite-olivine-chlorite schists, when veins occur containing talc-olivine-chlorite. Indeed, low H_2O activities seem to have been fairly common during the metamorphism of the higher grade ultramafics (TROMMSDORFF and EVANS, 1969a). Further study appears necessary before isograds can be mapped.

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